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The optical properties of particles deposited on a surface.

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by
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1. Outline of the research.

In recent years the detection and characterization of microscopic particles deposited on a surface has become an important problem both from the theoretical and the experimental point of view. The importance of a practical solution to this problem cannot be overemphasized. The final purpose of all the research that has been performed in this field is, indeed, the establishment of a standard technique, either theoretical or experimental or both, suitable for the detection and identification of the particles that may be deposited on sufaces that are supposed to be clean.

For homogeneous spherical particles a good method of detection and identification seem to be the study of the characteristic resonance peaks that appear in the plot of the extinction cross section vs. the wavelength. Nevertheless, it has been stressed by several researchers in the field that the extinction spectrum can undergo severe modifications when the particles are in the vicinity of an interface. This consideration is the basis of the research that we are going to describe, because we were able to find a simple method to use just these modifications to gain information on the morphology of the particles of interest.

2. Method of attack.

The method that we used to perform our research is based on image theory and is applicable to particles in the vicinity of a perfectly reflecting surface. Let us remark that the assumption of perfect reflectivity is satisfied to a high degree by almost any polished metallic surface. Under these conditions the problem of scattering by a particle in the vicinity of the surface has been proved to be quite equivalent to the prblem of the dependent scattering from the compound object that includes both the actual particle and its image provided that the exciting field is the superposition of the field that comes from the actual source and of the field that comes from the image source. the latter field coincides with the field that is actually reflected by the interface.

Under the preceding Contract DAJA45-93-C-0043 we performed a study of the scattering from anisotropic particles modeled as clusters of spheres and were able to show that, even when the particles are randomly oriented, thei anisotropy can be detected through a study of the polarized light that is scattered along the surface. Our present work is, in a sense, a continuation of the above

mentioned research.

Our basic idea is that, if the extinction spectrum is modified by the presence of a flat interface this must be due to the boundary conditions at the interface itself. Now, the reflection of a plane wave has seldom been studied in terms of multipole fields. Therefore we studied the reflection of the spherical multipole fields on a perfectly reflecting surface and were able to show that the condition of reflection imposes certain phase relations among the multipole amplitudes of the incident and of the reflected wave. Since, as stated above, the exciting field is just the superposition of the incident and of the reflected field we were able to show that several multipole emplitudes of the exciting field do vanish. It is just this vanishing that affects the resonance spectrum of a particle in the presence of a perfectly reflecting surface.

3. Sketch of the results.

The theory on which ve based our investigation is fully explained in the enclosed paper that has been submitted for publication to the Journal of the European Optical Society. Nevertheless it may be useful to summarize here the most important results of our calculatios on spheres and aggregated spheres.

When one considers homogeneos spheres it is possible to define a universal function that contains all the information on the polarization and on the direction of incidence of the exciting field. A study of this function allows one to predict the behavior of the resonance peaks for any spherical scatterer when the polarization and the angle of incidence are varied: the calculated spectra follow strictly the expected behavior.

We also made some calculations on the resonance spectra of aggregates composed of two identical mutually contacting spheres. In this case the lack of spherical symmetry prevents one from defining any useful function so that the calculations must be performed from scratch. A further difficulty stems from the fact that the multiple scattering processes among the spheres in the aggregate prevant the resonances to have a simple relation to the resonances of the component spheres. For this reason we considered only the resonances from aggregates of small spheres with a high refractive index. In this case, indeed, the only resonances that may occur are those with $l=1$. As a result we were able to perform an analysis of the extinction spectrum and to explain the behavior of the observed peaks when the reflecting surface is present. In spite of its simplicity the simple model that we dealt with gives the guidelines for the interpretation of the spectra of more complicated non-spherical particles.

4. List of publications.

F. Borghese, P. Denti, R. Saija, E. Fucile and O. I. Sindoni, "Morphology-dependent resonances of single and aggregated spheres in the presence of a perfetly reflecting surface," submitted to Journal of the European Optical Society.

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Morphology-dependent resonances of single and aggregated spheres in the presence of a perfectly reflecting surface

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The effect of the presence of a perfectly reflecting surface on the resonance spectra from particles is studied in the framework of the image theory. Due to the boundary conditions at the reflecting surface some of the resonances are expected to disappear. This suppression effect is studied with reference to single spheres and to binary aggregates of spheres and it is shown that a careful investigation of the scattered light as a function of the angle of incidence and of the polarization may give useful information on the morphology-dependent resonances both of spherical and non-spherical particles.

1. Introduction

The electromagnetic resonances are a well known feature of the extinction spectrum of a spherical scatterer. They occur in the plot of the extinction cross section vs. the size parameter as peaks that both the Mie theory[1] and the experiment[2–4] permit to relate to the multipole amplitudes of the incident wave. The analogy between the radial Helmholtz equation for spheres[5] and the radial Schrödinger equation for spherically symmetric systems led Johnson to formulate a comprehensive theory of the resonances both of homogeneous and of radially non-homogeneous spheres and to give, as a result, a number of useful formulas for the relevant parameters of each resonance.[6] Resonances may also occur in non spherical particles but they cannot be interpreted in the framework of the above mentioned theory neither for scatterers that are or can be modeled as aggregated spheres. In the latter case, indeed, the multiple scattering processes that occur among the spheres in the aggregate make the resonance spectrum to be in no way a mere superposition of the peaks that are due to the component spheres.[7,8] The resonance spectra proved useful to gain information on the morphology of the particles[9,2–4,10] so that several methods have been suggested to simplify their interpretation. For instance Li and Chýlek[11] suggested to modulate the observed resonances by illuminating the particle of interest with two counterpropagating plane waves. In turn, Johnson[12,13] suggested to study the scattering from a spherical particle coupled to a perfectly reflecting surface. In our opinion Johnson's procedure is rather promising although the presence of the reflecting surface requires considering the scattering from a binary cluster even when the actual scatterer is a single sphere. The image theory, indeed, states that the scattering from a sphere in the vicinity of a perfectly reflecting surface is quite equivalent to the dependent scattering from the compound object that includes both the actual particle and its image when illuminated by the superposition of the actual incident field and of the field that comes from the image source. Since the calculation of the dipendent scattering from aggregated spheres is a well established procedure[14] whose results are in excellent agreement with the available experimental data,[16,17] we were able to use the image theory to produce the full scattering pattern of an assembly of randomly oriented identical clusters in the vicinity of a metal surface for arbitrary direction and state of polarization of the incident wave.[15] In particular we found that the analysis of the polarization of the scattered light that propagates along the surface may provide useful information on the possible anisotropy of the particles.

The purpose of the present paper is to show that, as a result of the boundary conditions on a perfectly reflecting surface, some of the multipole amplitudes of the exciting field, that is defined as the superposition of the incident and the reflected field, vanish. As a consequence, some of the expected resonances may not appear in the observed spectrum: e. g. the peaks that are associated to a single vanishing multipole amplitude are bound to disappear. Using this suppression effect to interpretate the resonance spectra of particles of arbitrary shape is rather difficult,[18–24] however. Therefore, our investigation will deal with single spheres and with binary aggregates of spheres in the presence of a perfectly reflecting surface. In fact, although the resonance spectrum of aggregated spheres may be rather complicated, the guidelines for its interpretation can be given, at least in simple cases.

In Section 2 we revisit the reflection of a polarized plane wave on a perfectly reflecting surface in order to reformulate the problem in terms of spherical multipole fields.

In Section 3 we discuss the scattering from particles in terms of the transition matrix and give a general definition of the extinction cross section that applies even when a perfectly refelcting surface is present.

In Section 4 the resonance spectra from homogeneous spheres and from aggregates of two identical spheres are investigated on the basis of the theory of Sections 2 and 3.

A few concluding remarks are reported in Section 5.

2. Exciting field

In the framework of the image theory,[18] the exciting field is the superposition of the actual incident field and of the field that comes from the image source: the latter, in turn, coincides with the field that is reflected by the interface in the absence of any scatterer. Now, the reflection of a plane wave on a flat surface has seldom been dealt with in terms of multipole fields.[19,27] Therefore, we reformulate, in terms of multipole fields the relations that are dictated by the boundary conditions between the amplitude and the polarization of the incident and of the reflected wave. As a result we get the exciting field as an expansion in terms of spherical multipole fields that depend on the parameters of the incident wave only. At first we deal with an interface of general dielectric properties but we will specialize our results to the case of a perfectly reflecting interface when the need arises.

We consider a frame of reference whose cartesian axes are individuated by the unit vectors $\hat{\mathbf{u}}_x$, $\hat{\mathbf{u}}_y$ and $\hat{\mathbf{u}}_z$, and assume that the halfspace $z < 0$, the accessible half-space, is filled by a homogeneous medium of refractive index n while a different medium of refractive index n' fills the halfspace $z > 0$: thus the interface coincides with xy plane and its unit normal concides with $\hat{\mathbf{u}}_z$. The electromagnetic plane wave

$$\mathbf{E}_I = E_0 \hat{\mathbf{e}}_I \exp[i\mathbf{k}_I \cdot \mathbf{r}], \tag{1}$$

that propagates through the halfspace $z < 0$, is reflected by the interface into the plane wave

$$\mathbf{E}_R = E'_0 \hat{\mathbf{e}}_R \exp[i\mathbf{k}_R \cdot \mathbf{r}], \tag{2}$$

where $\hat{\mathbf{e}}_I$ and $\hat{\mathbf{e}}_R$ are the (unit) polarization vectors of the incident and of the reflected wave, respectively, $\mathbf{k}_I = nk\hat{\mathbf{k}}_I$ and $\mathbf{k}_R = nk\hat{\mathbf{k}}_R$ are the respective propagation vectors, and, as usual, $k = \omega/c$. The time dependence $\exp(-i\omega t)$ will be assumed throughout. To get the relations between the amplitudes and the polarizations of the incident and the reflected field one must also consider the transmitted plane wave

$$\mathbf{E}_T = E''_0 \hat{\mathbf{e}}_T \exp[i\mathbf{k}_T \cdot \mathbf{r}],$$

with $\mathbf{k}_T = n'k\hat{\mathbf{k}}_T$, that propagates through the halfspace $z > 0$, and impose the boundary conditions,

$$(E_0 \hat{\mathbf{e}}_I + E'_0 \hat{\mathbf{e}}_R + E''_0 \hat{\mathbf{e}}_T) \times \hat{\mathbf{u}}_z = 0, \tag{3}$$

across the interface. It is convenient to introduce two pairs of unit vectors $\hat{\mathbf{u}}_{I\eta}$ and $\hat{\mathbf{u}}_{R\eta}$ whose index $\eta = 1, 2$ distinguish wether they are parallel ($\eta = 1$) or perpendicular ($\eta = 2$) to the plane of incidence, i. e. to the plane that contains $\mathbf{k}_I$, $\mathbf{k}_R$ and the z axis. The orientation is chosen so that $\hat{\mathbf{u}}_{R2} \equiv \hat{\mathbf{u}}_{I2}$ and

$$\hat{\mathbf{u}}_{I1} \times \hat{\mathbf{u}}_{I2} = \hat{\mathbf{k}}_I, \quad \hat{\mathbf{u}}_{R1} \times \hat{\mathbf{u}}_{R2} = \hat{\mathbf{k}}_R.$$

Then Eqs. (1) and (2) can be rewritten as

$$\mathbf{E}_I = E_0 \sum_\eta (\hat{\mathbf{e}}_I \cdot \hat{\mathbf{u}}_{I\eta}) \hat{\mathbf{u}}_{I\eta} \exp[i\mathbf{k}_I \cdot \mathbf{r}] \tag{4}$$

$$\mathbf{E}_R = E'_0 \sum_\eta (\hat{\mathbf{e}}_R \cdot \hat{\mathbf{u}}_{R\eta}) \hat{\mathbf{u}}_{R\eta} \exp[i\mathbf{k}_R \cdot \mathbf{r}], \tag{5}$$

and the application of the boundary conditions, eq. (3), leads one to define the Fresnel coefficients F_η for the reflection of a plane wave with polarization along $\hat{\mathbf{u}}_\eta$. The expression of the coefficients F_η in terms of the angle ϑ_I between $\hat{\mathbf{k}}_I$ and $\hat{\mathbf{u}}_z$ is[25,26]

$$F_1 = \frac{n'^2 \cos\vartheta_I - n\sqrt{n'^2 - n^2 \sin^2\vartheta_I}}{n'^2 \cos\vartheta_I + n\sqrt{n'^2 - n^2 \sin^2\vartheta_I}}, \tag{6a}$$

$$F_2 = \frac{n \cos\vartheta_I - \sqrt{n'^2 - n^2 \sin^2\vartheta_I}}{n \cos\vartheta_I + \sqrt{n'^2 - n^2 \sin^2\vartheta_I}}, \tag{6b}$$

and their limiting value for the case of a perfectly reflecting surface is

$$F_\eta = (-)^{\eta-1}.$$

In terms of the Fresnel coefficients the relation between the components of the incident and of the reflected field is

$$E'_0 \hat{\mathbf{e}}_R \cdot \hat{\mathbf{u}}_{R\eta} = E_0 F_\eta \hat{\mathbf{e}}_I \cdot \hat{\mathbf{u}}_{I\eta},$$

i. e.

$$E'_0 = (-)^{\eta-1} E_0 F_\eta,$$

on account that

$$\hat{\mathbf{e}}_R \cdot \hat{\mathbf{u}}_{R\eta} = (-)^{\eta-1} \hat{\mathbf{e}}_I \cdot \hat{\mathbf{u}}_{I\eta},$$

and, as a result, the reflected plane wave, Eq. (5), can be rewritten as

$$\mathbf{E}_{\mathrm{R}} = E_0 \sum_\eta F_\eta (\hat{\mathbf{e}}_I \cdot \hat{\mathbf{u}}_{I\eta}) \hat{\mathbf{u}}_{R\eta} \exp[i\mathbf{k}_R \cdot \mathbf{r}]. \tag{7}$$

At this stage we recall that the multipole expansion of a vector plane wave of wavevector $\mathbf{K} = K\hat{\mathbf{K}}$ is[28,29]

$$\mathbf{E} = E_0 \hat{\mathbf{u}} \exp[i\mathbf{K} \cdot \mathbf{r}] = E_0 \sum_{plm} W^{(p)}_{lm}(\hat{\mathbf{u}}, \hat{\mathbf{K}}) \mathbf{J}^{(p)}_{lm}(\mathbf{r}, K),$$

where we define the spherical vector multipoles

$$\mathbf{J}^{(1)}_{lm}(\mathbf{r}, K) = j_l(Kr)\mathbf{X}_{lm}(\hat{\mathbf{r}}), \quad \mathbf{J}^{(2)}_{lm}(\mathbf{r}, K) = \frac{1}{K}\nabla \times j_l(Kr)\mathbf{X}_{lm}(\hat{\mathbf{r}}),$$

and the amplitudes

$$W^{(1)}_{lm}(\hat{\mathbf{u}}, \hat{\mathbf{K}}) = 4\pi i^l \hat{\mathbf{u}} \cdot \mathbf{X}^*_{lm}(\hat{\mathbf{K}}), \quad W^{(2)}_{lm}(\hat{\mathbf{u}}, \hat{\mathbf{K}}) = 4\pi i^{l+1} (\hat{\mathbf{K}} \times \hat{\mathbf{u}}) \cdot \mathbf{X}^*_{lm}(\hat{\mathbf{K}}).$$

In the preceding equations the superscripts 1 and 2 are the values of a parity index p that distinguishes the magnetic multipoles ($p = 1$) from the electric ones ($p = 2$) and the functions $\mathbf{X}_{lm}$ are vector spherical harmonics.[26] Using the notation and the phase conventions of Rose,[29] the latter functions can be defined in terms of the spherical harmonics $Y_{lm}(\hat{\mathbf{r}})$ as

$$\mathbf{X}_{lm}(\hat{\mathbf{r}}) = -\sum_\mu C(1, l, l; -\mu, m + \mu) Y_{l,m+\mu}(\hat{\mathbf{r}}) \boldsymbol{\xi}_{-\mu},$$

where

$$\boldsymbol{\xi}_0 = \hat{\mathbf{u}}_z, \quad \boldsymbol{\xi}_{\pm 1} = \mp \tfrac{1}{\sqrt{2}}(\hat{\mathbf{u}}_x \pm i\hat{\mathbf{u}}_y) \tag{8}$$

are the spherical basis vectors and the symbol C denotes the Clebsch-Gordan coefficients

$$C(1,l,l;\mp 1,m\pm 1)=\frac{\sqrt{l(l+1)-m(m\pm 1)}}{\sqrt{l(l+1)}},\quad C(1,l,l;0,m)=\frac{m}{\sqrt{l(l+1)}}.$$

Accordingly, the multipole expansions the incident and the reflected field are

$$\mathbf{E}_I = E_0 \sum_\eta (\hat{\mathbf{e}}_I \cdot \hat{\mathbf{u}}_{I\eta}) \sum_{plm} W^{(p)}_{I\eta lm} \mathbf{J}^{(p)}_{lm}(\mathbf{r}, nk) \tag{9a}$$

$$\mathbf{E}_R = E_0 \sum_\eta F_\eta (\hat{\mathbf{e}}_I \cdot \hat{\mathbf{u}}_{I\eta}) \sum_{plm} W^{(p)}_{R\eta lm} \mathbf{J}^{(p)}_{lm}(\mathbf{r}, nk), \tag{9b}$$

respectively, where we define

$$W^{(p)}_{I\eta lm} = W^{(p)}_{lm}(\hat{\mathbf{u}}_{I\eta}, \hat{\mathbf{k}}_I),$$
$$W^{(p)}_{R\eta lm} = W^{(p)}_{lm}(\hat{\mathbf{u}}_{R\eta}, \hat{\mathbf{k}}_R).$$

In order to relate the multipole amplitudes of the reflected wave, $W^{(p)}_{R\eta lm}$, to those of the incident wave, $W^{(p)}_{I\eta lm}$, we resort to the spherical basis, Eq. (8), to represent the vectors as

$$\mathbf{A} = \sum_\mu (-)^\mu (\mathbf{A}\cdot\boldsymbol{\xi}_\mu)\boldsymbol{\xi}_{-\mu} = \sum_\mu (-)^\mu A_\mu \boldsymbol{\xi}_{-\mu},$$

so that the dot product of any two vectors takes on the form

$$\mathbf{A}\cdot\mathbf{B} = \sum_\mu (-)^\mu A_\mu B_{-\mu},$$

and the spherical representation of any unit vector $\hat{\mathbf{v}}$ whose polar angles are ϑ and φ turns out to be[28]

$$\hat{\mathbf{v}} = \sum_\mu (-)^\mu v_\mu \boldsymbol{\xi}_{-\mu} = \sum_\mu (-)^\mu \sqrt{\frac{4\pi}{3}} Y_{1\mu}(\vartheta,\varphi)\boldsymbol{\xi}_{-\mu}.$$

Now, if ϑ_I and φ_I are the polar angles of the direction of incidence, we have

$$\begin{array}{lll} \hat{\mathbf{k}}_I \equiv (\vartheta_I, \varphi_I), & \hat{\mathbf{u}}_{I1} \equiv (\vartheta_I + \frac{\pi}{2}, \varphi_I), & \hat{\mathbf{u}}_{I2} \equiv (\frac{\pi}{2}, \varphi_I + \frac{\pi}{2}), \\ \hat{\mathbf{k}}_R \equiv (\pi - \vartheta_I, \varphi_I), & \hat{\mathbf{u}}_{R1} \equiv (\vartheta_I + \frac{\pi}{2}, \varphi_I + \pi), & \hat{\mathbf{u}}_{R2} \equiv (\frac{\pi}{2}, \varphi_I + \frac{\pi}{2}), \end{array}$$

so that it is an easy matter to see that the properties of the spherical harmonics under change of their arguments yield the following relation between the multipole amplitudes of the incident and of the reflected field

$$W^{(p)}_{R\eta lm} = (-)^{\eta+p+l+m} W^{(p)}_{I\eta lm}. \tag{10}$$

As a result the reflected field takes on the final form

$$\mathbf{E}_R = E_0 \sum_\eta F_\eta (\hat{\mathbf{e}}_I \cdot \hat{\mathbf{u}}_{I\eta}) \sum_{plm} (-)^{\eta+p+l+m} W^{(p)}_{I\eta lm} \mathbf{J}^{(p)}_{lm}(\mathbf{r}, nk), \tag{11}$$

that depends on the parameters of the incident wave only.

Equation (11) allows us to write the exciting field, that is the superposition of the incident and of the reflected field, as

$$\mathbf{E}_E = \mathbf{E}_I + \mathbf{E}_R = E_0 \sum_\eta (\hat{\mathbf{e}}_I \cdot \hat{\mathbf{u}}_{I\eta}) \sum_{plm} [1 + (-)^{\eta+p+l+m} F_\eta] W^{(p)}_{I\eta lm} \mathbf{J}^{(p)}_{lm}(\mathbf{r}, nk). \tag{12}$$

For a surface of general dielectric properties $|F_\eta| \neq 1$, so that the term within square brackets in eq. (12) never vanishes. However, for a perfectly reflecting interface $F_\eta = (-)^{\eta-1}$: therefore, when $[1-(-)^{p+l+m}]=0$, i. e. when $p+l+m$ is even, the corresponding multipole is not present in the exciting field.

3. Scattered field and extinction cross section

The field that is scattered by any particle embedded in a homogeneous medium of refractive index n can be expanded in a series of spherical vector multipoles

$$\mathbf{E}_{S\eta} = \sum_{plm} A_{\eta lm}^{(p)} \mathbf{H}_{lm}^{(p)}(\mathbf{r}, nk), \tag{13}$$

where the multipole fields $\mathbf{H}_{lm}^{(p)}$ are identical to the multipoles $\mathbf{J}_{lm}^{(p)}$ except for the substitution of the spherical Hankel functions of the first kind, $h_l^{(1)}(kr)$, to the spherical Bessel functions, $j_l(kr)$. The label η that is attached to $\mathbf{E}_{S\eta}$ and to the amplitudes $A_{\eta lm}^{(p)}$ recalls that the scattered field depends on the state of polarization of the incident wave. According to Waterman,[30] the multipole amplitudes of the exciting field are related to the amplitudes of the scattered field through the equation

$$A_{\eta lm}^{(p)} = -\sum_{p'l'm'} S_{lm,l'm'}^{(p,p')} W_{E\,\eta l'm'}^{(p')}, \tag{14}$$

where the quantities $S_{lm,l'm'}^{(p,p')}$ are the elements of the so called transition matrix, S, that accounts for the morphology (structure and scattering power) and the orientation of the particle. In the absence of any substrate the amplitudes $W_{E\,\eta l'm'}^{(p')}$ concide with those of the incident plane wave and S is the transition matrix of the actual scattering particle. However, when a perfectly reflecting surface is present the amplitudes of the exciting field, according to the preceding section, are

$$W_{E\eta lm}^{(p)} = [1 - (-)^{p+l+m}] W_{I\eta lm}^{(p)}, \tag{15}$$

and S should be the transition matrix appropriate to the compound object that includes both the actual particle and its image. In the latter case, according to Eq. (15), even a non vanishing incident amplitude $W_{I\eta lm}^{(p)}$ may yield a vanishing exciting amplitude $W_{E\eta lm}^{(p)}$ and thus affect the amplitudes of the scattered field up to the suppression of some of the characteristic resonance peaks. The possible occurrence of this suppression effect is, in principle, independent of the distance of the scatterer from the reflecting surface. Nevertheless, when the scatterer that we consider is sufficiently far from the surface the multiple scattering processes between the actual particle and its image become negligible and the scattered field becomes identical to the superposition of the fields that are scattered independently by the actual particle and by its image each illuminated by the exciting field.

We now need a definition of the extinction cross section that applies even in the presence of a flat interface. To this end we resort to the optical theorem that, according to van de Hulst,[1] can be proved by considering the field that is actually detected by an optical instrument. Let us recall that the scattering amplitude of any particle, $\mathbf{f}_\eta$, can be defined through the equation

$$\mathbf{E}_{S\eta} = \frac{\exp(inkr)}{r} E_0 \mathbf{f}_\eta(\hat{\mathbf{k}}_S, \hat{\mathbf{k}}_I), \tag{16}$$

provided the particle is at the origin and the distance of observation, r, is large. The scattering amplitude depends, in general, on the morphology as well as on the orientation of the scatterer with respect to the incident field and, once the amplitudes $A_{\eta lm}^{(p)}$ are known, its expression is

$$\mathbf{f}_\eta = \frac{1}{nk} \sum_{lm} (-i)^{l+1} \left[A_{\eta lm}^{(1)} \mathbf{X}_{lm}(\hat{\mathbf{k}}_S) + i A_{\eta lm}^{(2)} \hat{\mathbf{k}}_S \times \mathbf{X}_{lm}(\hat{\mathbf{k}}_S) \right]. \tag{17}$$

In terms of $\mathbf{f}_\eta$ the optical theorem reads

$$\sigma_{ext\,\eta} = -\frac{4\pi}{k^2}\mathrm{Im}[f_{\eta\eta}(\hat{\mathbf{k}}_S = \hat{\mathbf{k}}_I, \hat{\mathbf{k}}_I)], \tag{18}$$

where $f_{\eta,\eta'} = \mathbf{f}_\eta \cdot \hat{\mathbf{u}}^*_{S\eta'}$, and the index η recalls that, for an anisotropic scatterer, the cross section depends on the polarization. Now, according to van de Hulst,[1] Eq. (18) holds true even when a reflecting surface is present provided that the direction of observation be the direction of the reflected wave ($\hat{\mathbf{k}}_S \equiv \hat{\mathbf{k}}_R$): this is, indeed, the forward scattering direction when a reflecting surface is present. Nevertheless, one has to take account that, according to Section 2, when the incident wave is

$$\mathbf{E}_{I\eta} = E_0\hat{\mathbf{u}}_{I\eta}\exp(i\mathbf{k}_I \cdot \mathbf{r}),$$

i. e. when $\mathbf{E}_{I\eta}$ is either parallel ($\eta = 1$) or perpendicular to the plane of incidence ($\eta = 2$), the reflected wave is

$$\mathbf{E}_{R\eta} = E_0(-)^{\eta-1}\hat{\mathbf{u}}_{R\eta}\exp(i\mathbf{k}_R \cdot \mathbf{r}),$$

so that $\mathbf{E}_{R\eta}$ reverses its phase when the polarization of $\mathbf{E}_{I\eta}$ is perpendicular to the plane of incidence. Therefore, the total field that is detected by an optical instrument in the direction of reflection is

$$\mathbf{E}_{D\eta} = E_0[(-)^{\eta-1}\hat{\mathbf{u}}_{R\eta}\exp(i\mathbf{k}_R \cdot \mathbf{r}) + \frac{\exp(inkr)}{r}\mathbf{f}_\eta(\hat{\mathbf{k}}_R, \hat{\mathbf{k}}_I)],$$

and the optical theorem should read

$$\sigma_{ext\,\eta} = -\frac{4\pi}{k^2}\mathrm{Im}[(-)^{\eta-1}f_{\eta,\eta}(\hat{\mathbf{k}}_R, \hat{\mathbf{k}}_I)], \tag{19}$$

where

$$f_{\eta\eta}(\hat{\mathbf{k}}_R, \hat{\mathbf{k}}_I) = \frac{i}{4\pi nk}\sum_{plm}\sum_{p'l'm'} W^{(p)*}_{R\eta lm} S^{(p,p')}_{lm,l'm'} W^{(p')}_{E\eta l'm'}. \tag{20}$$

The phase factor $(-)^{\eta-1}$ in Eq. (19) does not appear explicitly in the expression given by Johnson[12] because this author considers only normal incidence and includes the correct phase in the expression for the reflected plane wave.

When the particle of interest is a sphere whose distance from the interface is sufficiently large that the interaction with its image need not be considered the exciting field is still the superposition of the incident and of the reflected field but the transition matrix is that appropriate to a single sphere. In this case S is diagonal,

$$S^{(p,p')}_{lm,l'm'} = \delta_{pp'}\delta_{ll'}\delta_{mm'}R^{(p)}_l, \tag{21}$$

and its elements are given by

$$R^{(p)}_l = \frac{(1+\overline{n}\delta_{p1})u_l(n_0k\rho)u'_l(nk\rho) - (1+\overline{n}\delta_{p2})u'_l(n_0k\rho)u_l(nk\rho)}{(1+\overline{n}\delta_{p1})u_l(n_0k\rho)w'_l(nk\rho) - (1+\overline{n}\delta_{p2})u'_l(n_0k\rho)w_l(nk\rho)}, \tag{22}$$

where

$$\overline{n} = \frac{n}{n_0} - 1, \quad u_l(x) = xj_l(x), \quad w_l(x) = xh^{(1)}_l(x).$$

The quantities $R^{(1)}_l$ and $R^{(2)}_l$ coincide with the Mie coefficients b_l and a_l, respectively, for the scattering by a homogeneous sphere of radius ρ and refractive index n_0 embedded in a homogeneous medium of refractive index n. Thus, by considering a sphere at least so distant from the interface, Eq. (20) can be rewritten as

$$f_{\eta\eta}(\hat{\mathbf{k}}_R, \hat{\mathbf{k}}_I) = \frac{i}{4\pi n k} \sum_{plm} W^{(p)*}_{R\eta lm} R^{(p)}_l W^{(p)}_{E\eta lm}. \tag{23}$$

On account that the elements $R^{(p)}_l$ are independent of m, it is meaningful to define the quantity

$$U^{(p)}_{\eta l} = \sum_m W^{(p)*}_{R\eta lm} W^{(p)}_{E\eta lm} \tag{24}$$

whose behavior as a function of the angle of incidence may give useful information for the interpretation of the resonance spectrum. In fact any resonance of a spherical scatterer that is associated to a vanishing $U^{(p)}_{\eta l}$ is bound to disappear.

4. Results and discussion

Before we go to discuss the results of our specific calculations it may be useful to recall a few facts about the electromagnetic resonances. For a spherical scatterer a resonance occurs when, with varying wavelength, one of the quantities $R^{(p)}_l$, Eq. (22), undergoes a fast change from a rather small value to a value of order unity. Since the quantities $R^{(p)}_l$ are the only non vanishing elements of the transition matrix of a sphere, Eq. (14) permits to associate each resonance to one and only one of the elements of the matrix S. For a nonspherical scatterer the occurrence of a resonance is no longer governed by the behavior of the diagonal elements of the transition matrix, because the off diagonal elements of S may play an important role. As a result, it is rather difficult to associate the observed resonances of a non spherical scatterer to a particular element of the transition matrix without a close examination of the behavior of all the relevant elements of S as a function of the wavelength. On the basis of the preceding considerations we decided to label the resonances both of spherical and of non spherical scatterers as magnetic and electric resonances according to whether they are associated to the magnetic part, $S^{(1,1)}_{lm,l'm'}$, or to the electric part, $S^{(2,2)}_{lm,l'm'}$, of the appropriate transition matrix; in this respect let us remark that for the scatterers that we are going to describe never occurred that a resonance should be associated with the mixed elements $S^{(1,2)}_{lm,l'm'}$ or $S^{(2,1)}_{lm,l'm'}$.

A. Single spheres

Our first step has been the calculation of the dependent scattering from the actual sphere and its image as a function of the distance d of the center of the actual sphere from the surface. By assuming that the homogeneous medium that fills the accessible half-space is the vacuum ($n = 1$) we found that, for a homogeneous spherical particle of radius ρ and (real) refractive index $n_0 = 3$, the multiple scattering processes between the particle and its image become negligibly small within the whole range of size parameter in which we are interested when $d = 10\rho$. Thus, if the sphere is at least so far from the interface we can use the formulas that we reported at the end of Section 3. In particular, for the quantity $U^{(p)}_{\eta l}$, Eq. (24), a straightforward calculation yields the result

$$U^{(p)}_{\eta l} = (-)^{\eta-1} 2\pi(2l+1) + \sum_m (-)^{\eta+p+l+m} W^{(p)*}_{I\eta lm} W^{(p)}_{I\eta lm}, \tag{25}$$

on account of Eqs. (10) and (15) and of the orthogonality properties of the W amplitudes.[31,32]

We report in Figs. 1 (a), (b), (c) and (d) the plots of $U^{(p)}_{\eta l}$ for $l \le 4$, $\eta = 1, 2$ and $p = 1, 2$, as a function of ϑ_I. We notice in Figs. 1 (a) and (b) that at $\vartheta = 0°$ all the $U^{(1)}_{1l}$ vanish for even l whereas all the $U^{(2)}_{1l}$ do vanish for odd l. This result was expected because when $\vartheta_I = 0°$ the

only non-vanishing amplitudes $W^{(p)}_{I\eta lm}$ are those with $m = \pm 1$. Therefore the above result is a consequence of the fact that the factor within square brackets in Eq. (10) reduces to $1 + (-)^{p+l}$. Although the preceding argument is based on the vanishing of the individual amplitudes $W^{(p)}_{E\eta lm}$ at $\vartheta_I = 0°$ the usefulness of the quantity $U^{(p)}_{\eta l}$ remains unaffected. In fact, the vanishing of $U^{(2)}_{1l}$ for $l = 2$ at $\vartheta_I = 45°$, Fig. 2 (b), is due to the sum over m in Eq. (24). The plots in Figs. 1 (c) and (d) show that the behavior of $U^{(p)}_{2l}$ is similar to that of $U^{(p)}_{1l}$. We remark that again all the $U^{(1)}_{2l}$ vanish at $\vartheta_I = 0°$ for even l whereas all the $U^{(2)}_{2l}$ vanish at the same incidence for odd l and, in particular, $U^{(2)}_{21}$ turns out to be identically zero: these features were expected on the ground of the structure of Eq. (25).

We report in Figs. 2 (a), (b) and (c) the quantity

$$\gamma_\eta = 2k\mathrm{Im}[(-)^{\eta-1} f_{\eta\eta}(\hat{\mathbf{k}}_R, \hat{\mathbf{k}}_I)],$$

for $\eta = 1$, for the sphere referred to above, in the presence of the reflecting surface, as a function of the size parameter $x = nk\rho$. The respective values of angle of incidence are $\vartheta_I = 0°, 45°, 70°$, the latter choice being suggested by the fact that, according to Figs. 1 (a) and (b), at this incidence none of the quantities $U^{(1)}_{1l}$ vanish although several of them assume a small value. The quantity γ_1 is as meaningful as $\sigma_{ext\,1}$ because it gives the extinction coefficient of a low density dispersion of identical scatterers[33]. In each of Figs. 2 we also report the plot of the quantity

$$\gamma = 2k\mathrm{Im}[f_{\eta\eta}(\hat{\mathbf{k}}_I, \hat{\mathbf{k}}_I)],$$

for the same sphere in the absence of any substrate. All the resonances were classified with the help of the well known formulas.[6,7,33] We stress that, in spite of the indices of f, the quantity γ is independent of the polarization, and that, for our purposes, γ_η is quite comparable to γ because both quantities refer to forward scattering, according to the discussion in Section 3.

The strict correspondence between the resonances that disappear and the vanishing of the respective $U^{(p)}_{1l}$ is so evident that, in our opinion, no further comment would be necessary. However, we call the attention of the reader on the simultaneous disappearence, in Fig. 2 (a), of the two resonances at $x = 1.3118$, that is associated to $p = 2$ and $l = 1$, and at $x = 1.437$, that is associated to $p = 1$ and $l = 2$: both peaks belong, in fact, to an odd value of $p + l$. The same mechanism explains also the simultaneous disappearence of the peaks at at $x = 2.2$ and at $x = 2.28$. The former peak is associated with $p = 2$ and $l = 3$ whereas for the latter peak $p = 1$ and $l = 4$: again, both peaks belong to an odd value of $p + l$. Even the resonance spectrum for $\eta = 2$ strictly follows the behavior of the corresponding $U^{(2)}_{\eta l}$ so that we resolved not to report the specific plot that, in spite of its significance, do not add any further information worth of a separate comment.

B. Binary clusters

The lack of a general theory applicable to non-spherical particles makes rather difficult an unambiguous classification of their resonances. According to Eq. (14), the lack of diagonality of the transition matrix prevents a meaningful definition of a function analogous to the quantity $U^{(p)}_{\eta l}$ that we defined above for single spheres. Even in the case of aggregated spheres the transition matrix is not a diagonal matrix so that there is no one-to-one association of the multipole amplitudes of the exciting field to those of the scattered field; nor there is a simple relation between the resonances of the component spheres and those of the aggregate as a whole. Nevertheless, Eq. (15) does not depend on the shape of the particles so that the vanishing of any of the amplitudes of the exciting field is expected to affect the resonances even of a non spherical particle. To show that this is, indeed, the case we resolved to investigate the resonance spectrum of an aggregate of two identical mutually contacting spheres of radius ρ both in the absence and in the presence of a perfectly reflecting surface. In this respect we recall that the procedure for the calculation of the

transition matrix of aggregated spheres is outlined in Ref. 15. Even in this case the medium that fills the accessible half-space was chosen to be the vacuum ($n = 1$); for the refractive index of the component spheres was assumed the unusually high value of $n_0 = 31.4$. According to Newton[33] and to our prevous experience[7], this choice makes the resonances both of the component spheres and of the aggregate as a whole to occur at so small values of $x = nk\rho$ that fully convergent values of the scattered field are obtained from Eq. (14) for $l = l' = 1$ only. As a result, the resonances of the aggregate as a whole can surely be associated to the multipole amplitudes with $l = 1$. In view of the anisotropy of any aggregate of spheres its orientation with respect to the incident field must be stated even when no reflecting surface is present. The axis of the binary aggregates that we consider was chosen to be parallel to the x axis of the frame of reference that we introduced in Section 2. The plane of incidence, in turn, always coincide with xz plane. Even for an aggregate of spherical scatterers of given orientation it is meaningful to introduce the quantity γ_η in strict analogy to the definition that we gave for spheres in Subsection A.

In Fig. 3 (a) we report, for both values of η, the quantity γ_η for the cluster referred to above in the absence of any reflecting surface whereas in Fig. 3 (b) we report γ_η for the case in which a reflecting surface is present and the axis of the aggregate lies at a distance $d = 10\rho$ from the surface. As in the case of the single spheres, indeed, our calculations show that at this distance the multiple scattering processes between the actual aggregate and its image are quite negligible. All the plots are reported as a function of $x = nk\rho$ and the angle of incidence is $\vartheta_I = 70°$.

Figures 4 (a) and (b) are identical to Figs. 3 (a) and (b), respectively, except that the angle of incidence is $\vartheta_I = 0°$.

On the whole, Figs. 3 and 4 present three peaks at $x_1 = 0.09629$, $x_2 = 0.09813$ and $x_3 = 0.10183$. Since for $l = 1$ the component spheres show a single resonance at $x = 0.1$, the plots in Figs. 3 and 4 confirm our previous statement in Section 1 that the multiple scattering processes within an aggregate produce resonances whose location cannot be related to the locations of the resonances of the component spheres.

The dependence of the resonanace spectrum of the aggregate on the polarization of the incident light can be understood only through a close examinantion of the features of the transition matrix and of the amplitudes of the exciting field. In this respect it is important to recall that the matrix S does not depend on the polarization so that the dependence on the polarization of the spectra in Figs. 3 and 4 are entirely due to the properties of the incident amplitudes $W^{(p)}_{I\eta lm}$.

Now, at x_1 the largest elements of S are $S^{(1,1)}_{1,\pm1;1,-1} \approx -S^{(1,1)}_{1,\pm1;1,1}$ so that a magnetic resonance ($p = 1$) is expected; at x_2 the leading elements are $S^{(2,2)}_{1,0;1,0}$ and $S^{(2,2)}_{1,\pm1;1,-1} \approx S^{(2,2)}_{1,\pm1;1,1}$ so that an electric resonance ($p = 2$) is expected; finally at x_3 the leading elements of S are $S^{(1,1)}_{1,0;1,0}$ and $S^{(1,1)}_{1,\pm1;1,-1} \approx S^{(1,1)}_{1,\pm1;1,1}$ thus suggesting that this resonance is a magnetic one. Nevertheless by comparing Figs. 3 and 4 one sees that not all the possible resonances actually occur. This is due to the dependence of the amplitudes $W^{(p)}_{I\eta lm}$ both on the polarization and on the angle of incidence ϑ_I.

As an example let us discuss the behavior of the resonance at x_2 that appears in Fig. 3 (a) for any choice of the polarization. According to Eq. (14) the implied amplitudes of the incident field are $W^{(2)}_{I\eta1,-1}$, $W^{(2)}_{I\eta1,1} = (-)^\eta W^{(2)}_{I\eta1,-1}$, $W^{(2)}_{I1,1,0} \neq 0$ and $W^{(2)}_{I2,1,0} = 0$. Therefore, when $\eta = 1$ the peak at x_2 belongs to $m = 0$ whereas for $\eta = 2$ it belongs to $m = \pm1$. When the reflecting surface is present, the implied amplitudes of the exciting field are $W^{(2)}_{E\eta1,\pm1} = 0$, $W^{(2)}_{E1,1,0} \neq 0$ and $W^{(2)}_{E2,1,0} = 0$. This is enough to explain why the peak at x_2 may appear in Fig. 3 (b) only for $\eta = 1$.

When $\vartheta_I = 0°$ the appropriate resonance spectra are those in Fig. 4. The behavior of the peak at x_2 is easily understood when one considers that at $\vartheta_I = 0°$ we have $W^{(2)}_{I1,1,0} = W^{(2)}_{E21,1,0} = 0$. Therefore a resonance can occur in Fig. 4 (a) for $\eta = 2$ only and cannot appear at all in Fig. 4 (b).

The behavior of the resonances at x_1 and at x_3 that appear in Figs. 3 and 4 can be easily understood through a quite similar analysis.

5. Conclusions

The results that we presented in Section 4 suggest that the presence of a perfectly reflecting surface may be a useful tool for the interpretation of the resonance spectra from scattering particles. However, the behavior of the spectra from spherical scatterers must be carefully distinguished from the behavior of the spectra from particles of more general shape.In fact, the diagonality of the transition matrix of spheres simplifies the association of the observed resonances to the multipole amplitudes of the exciting field: thus each resonance can easily be classified as electric or magnetic and attributed to the appropriate value of l. Further insight into the behavior of the resonances as a function of direction of propagation and of the polarization of the incident wave is supplied by the quantity $U_{\eta l}^{(p)}$, that, as we stressed in Section 4, can be defined only for spheres.

The interpretation of the resonances of particles of general shape is, on the contrary, rather difficult. In fact, since the transition matrix of such particles is non-diagonal, several elements of S can contribute to the same resonance. As a result, it is not easy to establish a correspondence between the observed resonances and the multipole amplitudes of the exciting field.

Even when the particles of interest are or can be modeled as clusters of spherical scatterers there is, in general, no evident relation between the resonances of the aggregate as a whole and those of the component spheres. In fact, the discussion in Section 4 on the resonance spectrum of binary clusters has been made possible only by choosing a small value for the radius and a rather high value for the refractive index of the component spheres. Due to our choice the resonances of the aggregate occur for $x \ll 1$ so that the convergence of the scattered field requires to include into the multipole expansions terms with $l = 1$ only. For realistic values of the refractive index the resonances of the cluster occur for higher values of x so that convergence of the scattered field requires to include into the multipole expansion, Eq. (13), terms with higher values of l: this, in turn, allows further resonances associated with higher values of l to appear. In spite of these difficulties, the simple example that we reported in Section 4 suggests that a careful analysis as a function of ϑ_I and of the polarization both in the presence and in the absence of a reflecting surface may give useful information for the interpretation of the resonance spectra of non-spherical particles.

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Fig. 1. Plot of the quantity $U_{\eta l}^{(p)}(\vartheta_I)$ for $l \leq 4$. In (a) $p = 1$ and $\eta = 1$; in (b) $p = 2$ and $\eta = 1$; in (c) $p = 1$ and $\eta = 2$; in (d) $p = 2$ and $\eta = 2$. In this paper we adopted the convention that $p = 1, 2$ classifies the multipoles as magnetic and electric, respectively; in turn $\eta = 1, 2$ indicates that the polarization is parallel or orthogonal to the plane of incidence, respectively.

Fig. 2. γ_η (solid curve) and γ (dotted curve) for a homogeneous sphere of radius ρ and refractive index $n_0 = 3$ as a function of $x = nk\rho$ for $\eta = 1$. The medium that fills the accessible half-space ia assumed to be the vacuum ($n = 1$). The distance between the center of the sphere and the reflecting surface is $d = 10\rho$. The angle of incidence is $\vartheta_I = 0°$ in (a), $\vartheta_I = 45°$ in (b) and $\vartheta_I = 70°$ in (c). The resonances are classified according to the scheme $(p, l)_n$, where n distinguishes different resonances with the same value of p and l.

Fig. 3. γ_η for the aggregate of two identical mutually contacting spheres of radius ρ and refractive index $n_0 = 31.4$ as a function of $x = nk\rho$. The medium that fills the accessible half-space ia assumed to be the vacuum ($n = 1$). The axis of the aggregate lies in the xz plane and is parallel to the x axis. The plane of incidence coincides with the xz plane. The angle of incidence is $\vartheta_I = 70°$. The solid and the dotted curves refer to polarization parallel and orthogonal to the plane of incidence, respectively. In (a) no reflecting surface is present, whereas in (b) a reflecting surface that coincide with the xy plane is present and the distance between the axis of the aggregate and the surface is $d = 10\rho$. We notice that the spike at $x = 0.09813$ in Fig. 3 (a) appears for both choices of the polarization, although this is not easily discernible on the scale of the figure.

Fig. 4. Same as Figure 3 except that the angle of incidence is now $\vartheta_I = 0°$.

Fig. 1. Plot of the quantity $U_{\eta l}^{(p)}(\vartheta_I)$ for $l \leq 4$. In (a) $p = 1$ and $\eta = 1$; in (b) $p = 2$ and $\eta = 1$; in (c) $p = 1$ and $\eta = 2$; in (d) $p = 2$ and $\eta = 2$. In this paper we adopted the convention that $p = 1, 2$ classifies the multipoles as magnetic and electric, respectively; in turn $\eta = 1, 2$ indicates that the polarization is parallel or orthogonal to the plane of incidence, respectively.

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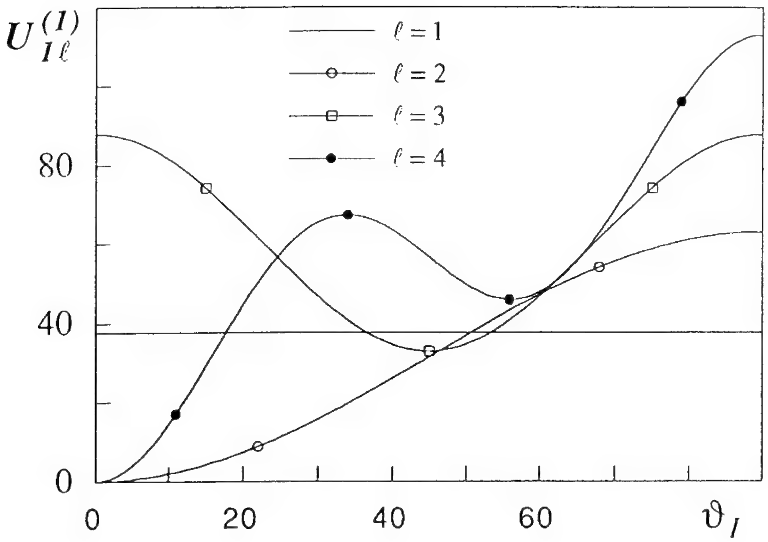
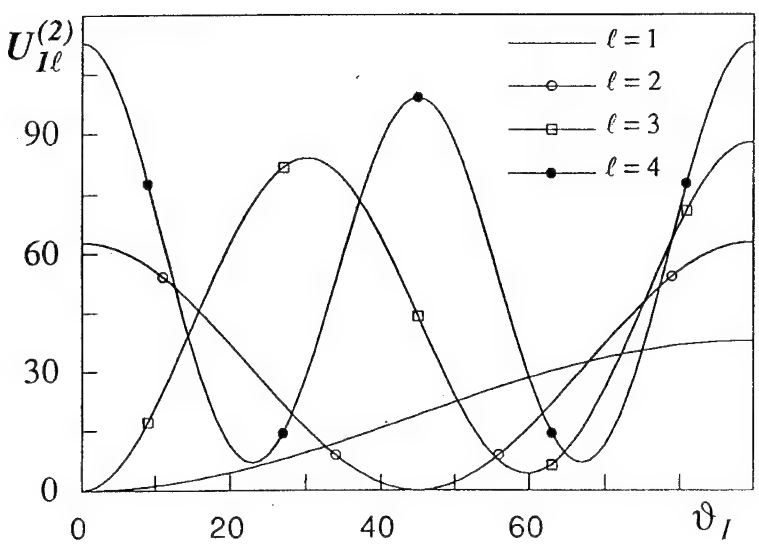
Fig. 4. Same as Figure 3 except that the angle of incidence is now $\vartheta_I = 0°$.

Fig. 1



$U^{(1)}_{1\ell}$
$\ell = 1$
$\ell = 2$
$\ell = 3$
$\ell = 4$
80
40
0
0
20
40
60
ϑ_l

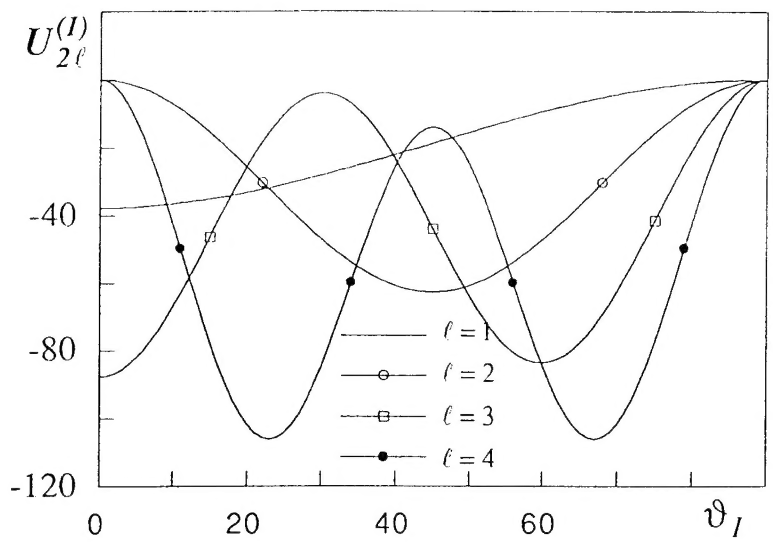
a)



$U^{(2)}_{1\ell}$
$\ell = 1$
$\ell = 2$
$\ell = 3$
$\ell = 4$
90
60
30
0
0
20
40
60
ϑ_l

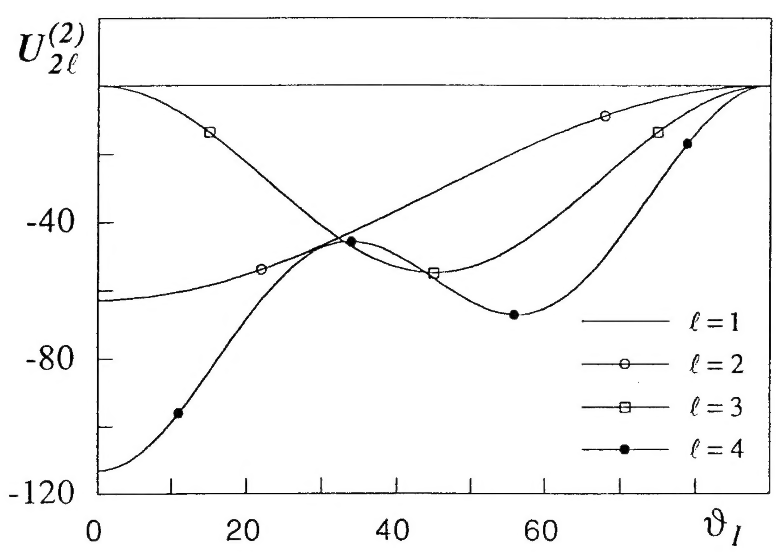
b)

Fig 1.



$U_{2\ell}^{(1)}$
-40
-80
-120
0
20
40
60
ϑ_I
$\ell = 1$
$\ell = 2$
$\ell = 3$
$\ell = 4$

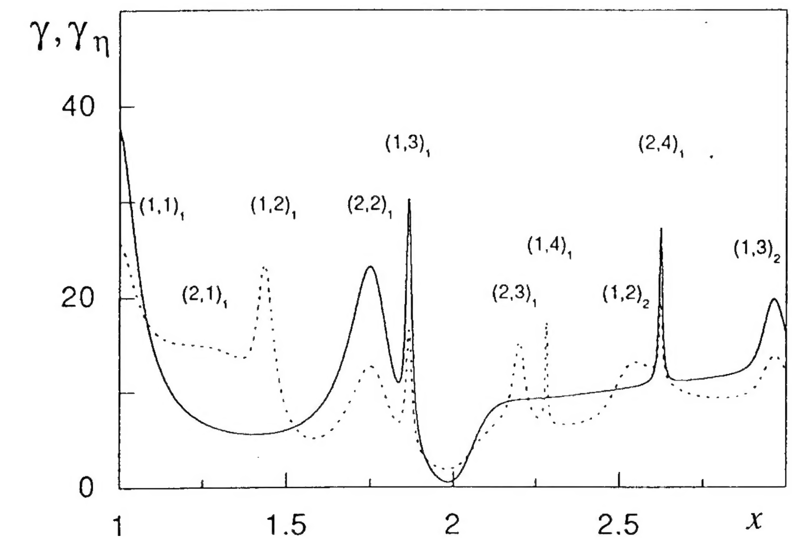
c)



$U_{2\ell}^{(2)}$
-40
-80
-120
0
20
40
60
ϑ_I
$\ell = 1$
$\ell = 2$
$\ell = 3$
$\ell = 4$

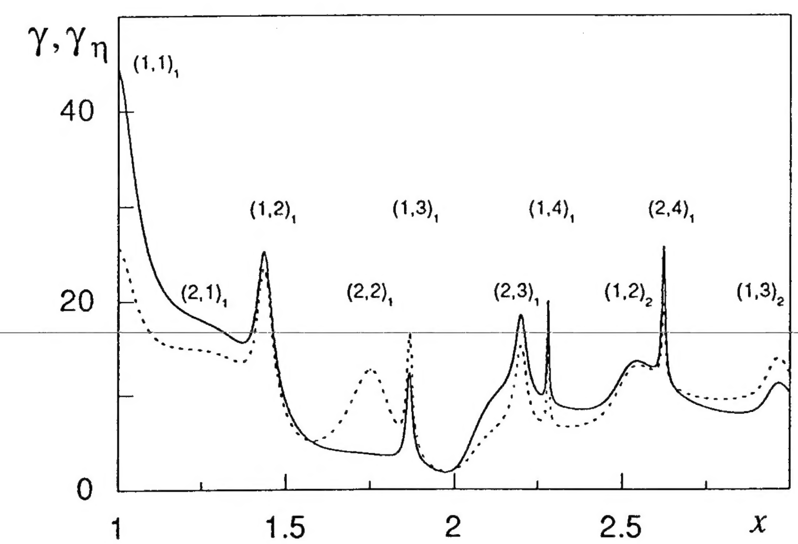
d)

Fig 2.



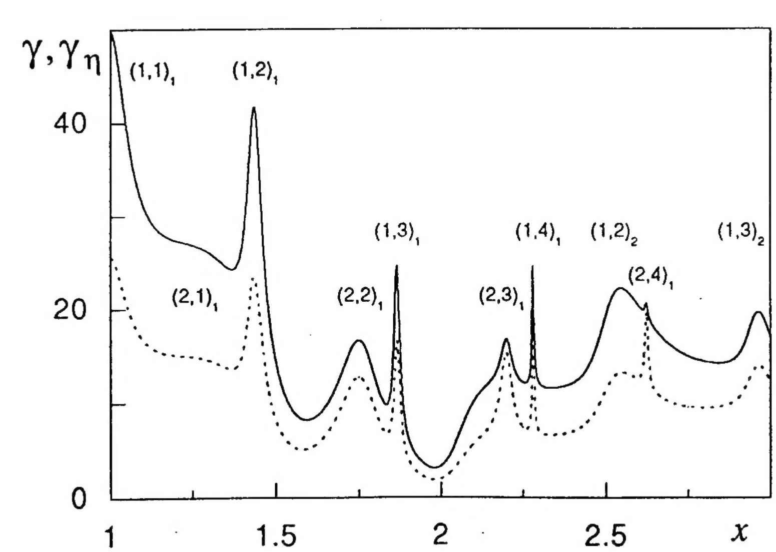
γ, γ_η
40
20
0
1
1.5
2
2.5
x
$(1,1)_1$
$(2,1)_1$
$(1,2)_1$
$(2,2)_1$
$(1,3)_1$
$(2,3)_1$
$(1,4)_1$
$(1,2)_2$
$(2,4)_1$
$(1,3)_2$

a)



γ, γ_η
40
20
0
1
1.5
2
2.5
x
$(1,1)_1$
$(2,1)_1$
$(1,2)_1$
$(2,2)_1$
$(1,3)_1$
$(2,3)_1$
$(1,4)_1$
$(1,2)_2$
$(2,4)_1$
$(1,3)_2$

b)



γ, γ_η
40
20
0
1
1.5
2
2.5
x
$(1,1)_1$
$(1,2)_1$
$(2,1)_1$
$(2,2)_1$
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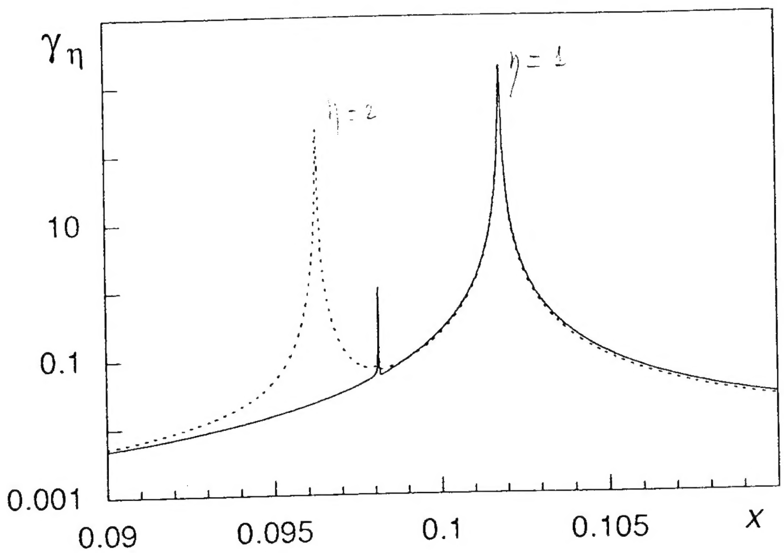
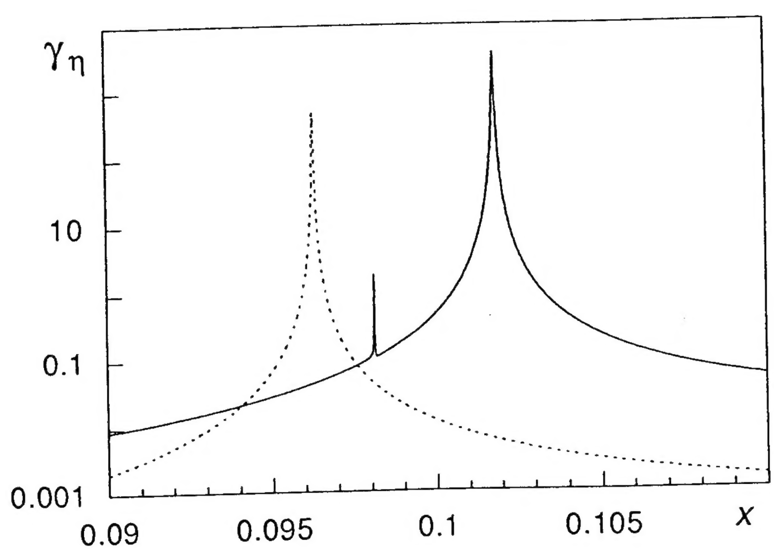
$(2,4)_1$
$(1,3)_2$

c)



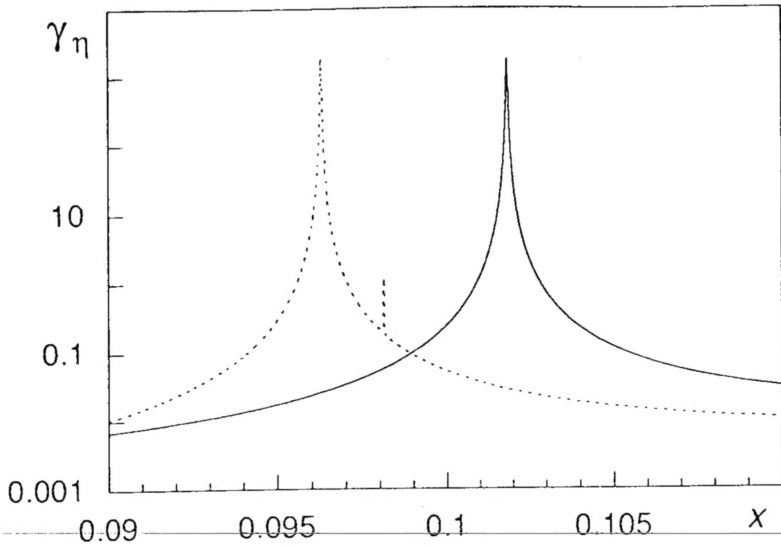
γ_η
$\eta = 1$
$\eta = 2$
10
0.1
0.001
0.09
0.095
0.1
0.105
x

F3a

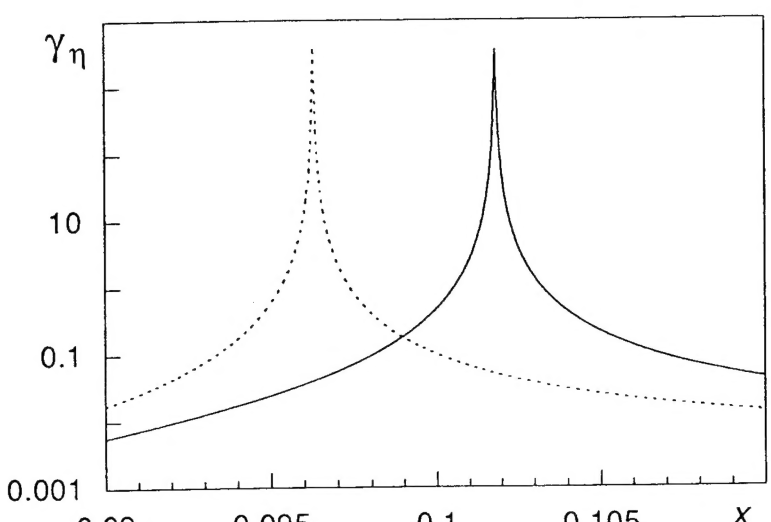


γ_η
10
0.1
0.001
0.09
0.095
0.1
0.105
x

F3b



F4a



F4b